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14. ABSTRACT

We have demonstrated methods to apply forces due to the scattering of photons, which are commonly used to manipulate atoms, to the more complex system of diatomic molecules. This has made it possible to use lasers to deflect, slow, and cool a beam of diatomic molecules. Possible applications of laser-cooled molecules range from quantum information processing and quantum simulation to the study of quantum chemical dynamics to precision measurements of fundamental symmetries. In the course of this work, we also made a detailed characterization of a

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Report Title

Trapping and cooling of polar molecules

ABSTRACT

We have demonstrated methods to apply forces due to the scattering of photons, which are commonly used to manipulate atoms, to the more complex system of diatomic molecules. This has made it possible to use lasers to deflect, slow, and cool a beam of diatomic molecules. Possible applications of laser-cooled molecules range from quantum information processing and quantum simulation to the study of quantum chemical dynamics to precision measurements of fundamental symmetries. In the course of this work, we also made a detailed characterization of a new type of cryogenic molecular beam source, which made the observation of laser cooling possible and also may have wider applications in chemical physics; and we developed a simple method for laser frequency locking, with reconfigurable parameters, based on commercial Field Programmable Gate Array (FPGA) digital circuits.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
02/26/2013	6.00	G Yang, J F Barry, E S Shuman, M H Steinecker, D DeMille. A low-cost, FPGA-based servo controller with lock-in amplifier, Journal of Instrumentation, (10 2012): 10026. doi: 10.1088/1748-0221/7/10/P10026
08/17/2012	4.00	L. Hunter, S. Peck, A. Greenspon, S. Saad Alam, D. DeMille. Prospects for laser cooling TlF, Physical Review A, (01 2012): 0. doi: 10.1103/PhysRevA.85.012511
08/17/2012	5.00	E. Norrgard, D. DeMille, J. Barry, E. Shuman. Laser Radiation Pressure Slowing of a Molecular Beam, Physical Review Letters, (03 2012): 0. doi: 10.1103/PhysRevLett.108.103002
08/29/2011	1.00	E. S. Shuman, D. DeMille, J. F. Barry. A bright, slow cryogenic molecular beam source for free radical, Phys. Chem. Chem. Phys., (06 2011): 0. doi:
08/29/2011	2.00	E. S. Shuman, J. F. Barry, D. DeMille. Laser cooling of a diatomic molecule, Nature, (9 2010): 820. doi: 10.1038/nature09443

TOTAL: 5

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received	Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

"Laser Cooling of a Diatomic Molecule", presented at the Symposium on Physics and Chemistry of Coherently Controlled Quantum Systems, Institute for Molecular Spectroscopy, Inuyama, Japan, March 2010

"Laser Cooling of a Diatomic Molecule", presented at the workshop on Coherence in Ultracold Molecular Physics, University of British Columbia, May 2010

"Direct Laser Cooling of a Diatomic Molecule", presented at the APS DAMOP meeting, May 2010

"Progress Towards Laser Cooling and Trapping Strontium Monofluoride from a Cryogenic Beam Source", presented at the APS DAMOP meeting, May 2010

"Laser Cooling of a Diatomic Molecule", presented at the Taiwan International Workshop on Ultracold Atoms/Molecules, June 2010

EUROQUAM Conference, Ischgl, Austria, Sept. 2010

MURI Ultracold Molecules Workshop, Boulder, CO Oct. 2010

APS DAMOP Meeting, Atlanta, GA, June 2011

ITAMP Workshop on Fundamental Symmetries and Ultracold Molecules, Cambridge, MA, Sept. 2011

CLEO/QELS Conference, Baltimore, MD, May 2011
Optical Society of America/Frontiers in Optics/Laser Science Conference, Rochester, NY, Oct. 2011

Physics of Quantum Electronics, Snowbird, Utah, USA, Jan. 2012.

ITAMP Workshop on Research Frontiers in Ultra-Cold Atoms and Molecules: Unequal Mass Mixtures and Dipolar Molecules, Cambridge, MA, May 2012

Number of Presentations: 12.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received		Paper
02/27/2013	7.00	E. Shuman, J. Barry, D. DeMille. Transverse laser cooling of a strontium monofluoride molecular beam, International Conference on Laser Spectroscopy 2011. 2011/05/29 00:00:00, . : ,
TOTAL:		**1**

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Paper

TOTAL:

Patents Submitted

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
David Glenn	0.01	
Lucas Willis	0.01	
Eric Norrgard	0.01	
David Naylor	0.07	
John Barry	0.34	
Eustace Edwards	0.01	
FTE Equivalent:	**0.45**	
Total Number:	**6**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
Edward Shuman	0.60
FTE Equivalent:	**0.60**
Total Number:	**1**

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
David DeMille	0.01	
FTE Equivalent:	**0.01**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED	Discipline
Yang Ge	0.01	Physics
Chris Zeng	0.02	Physics
FTE Equivalent:	**0.03**	
Total Number:	**2**	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 1.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 1.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 1.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 1.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

NAME
Total Number:

Names of personnel receiving PHDs

NAME	
David Glenn	
Total Number:	**1**

Names of other research staff

NAME	PERCENT_SUPPORTED
Matt Steinecker	0.01
FTE Equivalent:	**0.01**
Total Number:	**1**

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

Our progress has come on several different fronts:

1. Demonstration of optical force via radiative cycling in SrF molecules. We demonstrated the ability to create an effective cycling transition in SrF molecules, using only 2-3 diode lasers (with the second and third lasers for repumping from excited vibrational levels) and a static magnetic field (for remixing out of dark Zeeman sublevels). With two lasers, we demonstrated up to 100 photon scattering events with loss too small to observe (<5%). The number of scattered photons was determined both by observing laser-induced fluorescence, and by observing the deflection of an SrF beam by the optical force due to photon scattering. In the course of these measurements, we understood that the dark Zeeman and vibrational sublevels in the ground state of our cycling scheme (using a N=1 <--> N'=0 P-branch rotational line) leads to an effective reduction in the photon scattering rate by a factor of several, compared to a true two-level system such as an atom. This crucial realization must be taken into account in any molecular laser cooling scheme; it is inconvenient, but not crippling.

2. Demonstration of laser cooling of SrF molecules. By extending the methods used to demonstrate optical deflection, we next demonstrated the transverse cooling of an SrF beam. We first observed Doppler cooling, in good agreement with theoretical expections. We also observed, unexpectedly, a strong Sisyphus cooling force under certain conditions. We determined that the origin of this force is the same as that observed over 20 years ago in atoms cooled on a similar transition with dark Zeeman sublevels. The ultimate temperature achieved with both methods was limited by the interaction time of molecules with the lasers, but was still the lowest achieved with any direct molecular cooling method (~5 mK for Doppler cooling and ~300 uK for Sisyphus cooling).

3. Careful characterization of the cryogenic molecular beam source. Our work on optical cycling/deflection and laser cooling used a new type of cryogenic molecular beam source. Earlier work in our group and the group of John Doyle at Harvard had described some properties of these beams, but we have now made much more careful and complete measurements. Briefly, the source uses a cryogenically-cooled cell filled with He gas at ~4K as a coolant for the molecules of interest. Molecules are injected into the cell via laser ablation from a solid target in the cell, then extracted into a beam via a hole in the cell. We find that in a certain regime of helium flow rates through the cell, the beam has some unusual and highly favorable properties. In particular, for mildly supersonic helium flows through the exit aperture, the beam of SrF molecules has a low divergence (nearly 10 times smaller than for a typical molecular beam) and very low rotational temperature (<1 K) as well as fairly low velocity (200 m/s maximum, and as low as 120 m/s). All these features are maintained while achieving total flux that is substantially larger than the best other sources for free radical and refractory species. We are preparing a paper on this work.

4. Development of a new electronic system for locking the frequency of tunable lasers. This is based on FPGA (field programmable gate array) technology, which is a very flexible and fast form of digital signal processing. We use inexpensive, commercially-available FPGA evaluation boards with built-in analog-to-digital and digital-to-analog converters to emulate the function of standard analog circuitry for laser frequency servo locking—including a lock-in amplifier and two lines of PI servo feedback. The evaluation board includes a convenient user interface (LCD display plus rotary knobs, pushbuttons, and switches) that makes it possible for a user to change all system parameters (such as filter cutoff frequencies, amplifier gains, etc.) on the fly.

5. Demonstration of laser longitudinal slowing of a beam SrF molecules. We successfully slowed the velocity v of the SrF beam from v~130 m/s (peak of the distribution) to v~50 m/s (peak), with a substantial fraction of the population having v<25 m/s. The latter velocity should be sufficiently for loading molecules into a variety of traps, including a magneto-optic trap. Note that, because we must apply multiple RF sidebands to our laser in order to address the spin-rotation and hyperfine substructure of the SrF molecules, our conditions are similar to those used in atomic “white-light slowing” protocols. In analogy with these long-known results, the entire range of the velocity distribution is slowed but there is little longitudinal cooling.

Because our data, taken at face value, appears to show a factor of 2-3 loss in molecular flux when the largest slowing is observed, we carefully investigated a variety of possible loss mechanisms during the slowing. By probing the population in all conceivable “reservoir” levels where molecular population could be trapped, we find that there is negligible leakage out of the engineered cycling transition (including two vibrational repump lasers) to other rotational or vibrational states. (Note that this is in agreement with our estimates of the leakage out of the cycle, and is quite remarkable for a molecule given that the slowing data indicates that a typical molecule has scattered \gtrsim 10^4 photons.) Rather, we believe the apparent loss to be an artifact of our detection method. In particular: as the molecules are slowed longitudinally but not transversely, the beam divergence increases and fewer slow molecules hit our finite-size detection region. In addition, spontaneous emission of photons in random directions during each scattering event heats the transverse velocity of the slowed molecules. This further amplifies the effect of apparent loss due to divergence. A Monte Carlo simulation of molecular trajectories in our beam, with and without slowing, indicated that if the loss due to divergence is accounted for, then very few molecules are lost and a very large number have been slowed to velocities v< 25 m/s.

Such a slow and cold beam should be suitable for loading a magneto-optic trap of SrF molecules.

6. In a side project, we collaborated with Larry Hunter (Amherst College) to evaluate the suitability of a new molecular species, thallium monofluoride (TlF), for laser cooling. Hunter has measured the lifetime ($\tau \approx 100$ ns) and Franck-Condon factors for decay of the $X\ ^1\Sigma$ – $B\ ^3\Pi_1$ transition in TlF. Both agree with simple estimates, and indicate that TlF is a likely candidate for laser cooling. This species is chemically quite different from other species now being used for molecular laser cooling (i.e., it has a closed-shell ground state rather than an open shell), and in addition has possible applications to precision measurements of fundamental symmetries (in particular, the time-reversal odd “Schiff moment”, conceptually similar to the electric dipole moment of a nucleus).

7. We began a study of the possibility to use stimulated laser forces rather than spontaneous scattering forces to slow a molecular beam. We performed some calculations on a model system (with structure similar to that of SrF, but simpler) using the “bichromatic force” (essentially, applying counter-propagating laser beams, each consisting of two frequency components). In this model we see that it should be feasible to apply forces at least ~10 times greater than the spontaneous scattering force, using this method. This would make it possible to slow beams of molecules with less favorable cycling properties than SrF. In addition, data from atoms indicates that the bichromatic force is in fact capable of cooling in addition to slowing, though the mechanism that makes this possible (despite the fact that the forces all seem to be conservative) is not understood. Hence even molecular cooling may benefit from this technique. We also devised several new, different, and plausible methods for stimulated-force beam slowing, including near-resonant optical Stark deceleration and forced Sisyphus cooling. We showed that quite generally, these methods are all most effective when a tunable laser with moderately long pulse duration (~100 microsec) and high pulse energy (~100 mJ) can be used. We have begun construction of such a laser.

8. In the final months of this grant, our primary activity was the near-complete redesign and reconstruction of our SrF apparatus, to allow us to deliver a large flux of slow SrF molecules to an ultra-high vacuum region suitable for a magneto-optic trap (MOT) of SrF. The initial goal was to implement simultaneous transverse cooling and radiative slowing of the beam. We had simulated the effect of this, and predicted a very large increase in useful flux (slow enough to capture in a MOT). Unfortunately, our data showed a much smaller increased on application of 1D transverse cooling, and in addition a diminution of the slowing force when transverse cooling is applied. We now understand this as due to competition between these processes: when a molecule is scattering a photon from one of the laser beams, it cannot be scattering from the other beam. Hence, our expectations for available flux for MOT loading had to be significantly lowered. Nevertheless, we have continued construction of the full slowing and trapping apparatus, and it is now nearly complete.

9. In the meantime, we devised a different method that promises to deliver a large gain in available flux of slow molecules for MOT loading. The primary difficulty is due to the fact that the molecular beam’s divergence causes loss of molecules into a fixed volume downstream; and in addition, as the molecules are slowed their divergence increases. Transverse cooling can combat this, but as documented in our earlier work, scattering rates and slowing forces are small in molecules due to the multiple internal energy levels involved in the optical cycling process. Hence, in our initial attempts at simultaneous transverse cooling and slowing, we found that either a) the transverse cooling force, diminished by competition with the slowing force, was too small to be effective before the molecules were slowed; or b) by the time the molecules were slow enough that transverse cooling could be effective, they would have diverged out of the useful volume. Hence, we sought a method that could transversely confine molecules while they are being slowed, after which they can be transversely cooled with high efficiency (since the residence time of slow molecules in a transverse cooling region of finite is much longer). We subsequently realized that the necessary features can be provided by a microwave field, nearly resonant with a rotational transition in the molecule, with power enhanced in a near-resonant cavity. The concept is similar to that described in our old paper on the theory of microwave trapping of molecules [D. DeMille, D. R. Glenn, and J. Petricka, Eur. Phys. J. D 31, 375 (2004)], and recently demonstrated elsewhere [S. Merz et al., Phys. Rev. A 85, 063411 (2012)] for guiding and deceleration of a molecular beam. Here, we will guide molecules in the rotational N=1 state used in the laser cooling cycle, using microwaves tuned to the blue of the N=1 <--> N=2 rotational transition at ~30 GHz. Using the TE01 mode of a cylindrical cavity, N=1 molecules are confined to the center of the cylinder with a trapping force that (for carefully chosen frequency detuning) is nearly independent of the spin-rotation/hyperfine/Zeeman sublevel of the molecule. We have obtained a high-power (~40 W) microwave amplifier, and designed and tested a suitable cavity and power delivery scheme. Based on our results so far, we anticipate being able to capture and guide 100-1000 times more slow molecules than in our current apparatus. We note that nominally similar methods for guiding (e.g. with electrostatic or magnetic guides) are incompatible with laser slowing, for a variety of reasons. Most importantly, they are only effective on a fraction of molecular sublevels and in fact lead to “anti-guiding” of at least half of the subelevels, while laser slowing rapidly redistributes population between sublevels. In addition, the trivial ability to rapidly switch on and off microwave power in a cavity makes it possible to alternate between guiding and slowing forces, with relative duty cycle of each chosen to optimize the delivery of useful flux to the trap. Once the molecules emerge from the guide, they can be transversely cooled if necessary for optimal trap loading; here transverse cooling can be very effective over a short distance, since the molecules will be moving very slowly already.
A paper on these findings is in preparation.

10. Finally, we also began construction of an apparatus to study magneto-optic trapping of Rb atoms on their D1 line. This system bears many features in common with the MOT of SrF—in particular, the existence of multiple dark Zeeman sublevels in the ground state, and the absence of an obvious mechanism for net restoring forces in a MOT on this transition. We believe a better understanding of the mechanisms at play in a D1 line MOT will be very useful as we begin work on the SrF MOT. One previous paper has reported successful operation of a MOT on the D1 line of Na atoms, with the number of trapped atoms reported as similar to that in a standard D2 line MOT where the trapping mechanism is well understood. However, this paper provided no explanation for the trapping mechanism, and only minimal detail about the resulting conditions of the trapped atoms, which experimental parameters were important, etc. Our plan is to study these in more detail with Rb in this new apparatus.

Technology Transfer